

On the Equivalence between Mode-Coupling Theory and Replica Approach for Super-Cooled Liquids

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We show that the replica approach to glassy dynamics provides, in spite of its static nature, a characterization of critical dynamics in the β -regime of super-cooled liquids that is equivalent to the one of Mode-Coupling-Theory, both qualitatively and quantitatively. The nature and extent of this equivalence is discussed in connection to the main open problems of the current theory.

It is nowadays well established that the Mode-Coupling-Theory (MCT) yields an accurate description of the early stages of the dynamical slowing down in super-cooled glass-forming liquids [1]. One of its main successes is the prediction that the slow relaxation has a two-step nature with time correlators developing plateaus that are expected to diverge at some critical temperature characterized by a transition to a glass phase.

The main drawback of the theory is that the critical temperature is actually significantly larger than the one of the glass-transition in numerical simulations and it seems rather to mark a dynamical cross-over [2]. Nevertheless various quantities, like the so-called non-ergodicity parameter (essentially the height of the plateau of the correlators), compare very well with the numerical data. Furthermore the theory provides a detailed set of predictions for the behavior of the critical correlators and the dynamical exponents [3] that reproduce quite well the numerical data [4–6]. For these reasons many believe that the inclusion of some sort of corrections to the theory, possibly taking into account finite-dimensional effects leading to some sort of hopping processes, could extend MCT down to the laboratory glass transition. Unfortunately due to the uncontrolled nature of Sjogren's approximation [7] which is at the heart of standard MCT [8] it is not clear how to compute systematically corrections to it.

On the other hand it has been discovered by Kirkpatrick, Thirumalai and Wolynes [9] that a certain class of mean-field spin-glass models exhibits the same dynamical features of MCT. This result is important for various reasons. It shows that MCT works for systems that are completely different at the microscopic level from a liquid hinting that it might have some universal features. Furthermore it allows to include, at least phenomenologically, finite-size effects in the theory. Indeed critical dynamics in these models can be tracked back to the splitting of the equilibrium state into an exponential number of metastable states and the use of nucleation arguments on this mean-field picture has led to the formulation of the so-called Random-First-Order-Theory of the glass transition [9, 10]. Another important consequence is that some features of critical dynamics of these models, namely the critical temperature and the non-ergodicity parameter, can be computed without solving explicitly the dynamics by means of an essentially a static

approach: the replica method. This observation has led to the application of the replica method directly in the context of liquid theory, leading to a novel set of quantitative predictions for the non-ergodicity parameter and for the critical temperature [11].

In the replica approach one is typically concerned with a replicated order parameter q_{ab} that describes the appropriate microscopic two-point correlation (*i.e.* density-density in liquids or spin-spin in spin-glasses) between two identical replicas of the system. Using standard statistical mechanics procedures one derives a (usually approximate) equation for q_{ab} and the dynamical glass transition is characterized by the abrupt appearance of a novel solution of the equation characterized by a value of q_{ab} different from the value corresponding to the high temperature phase (say the liquid or paramagnetic phase depending on the system considered). The solution depends on the number m of replicas considered and for technical reason one should take the limit $m \rightarrow 1$ in order to obtain the correct result. In the structural glass context it is customary to use methods based essentially on the Hyper-netted-chain (HNC) approximation [11, 12] that give predictions for the non ergodicity parameter $f(q)$ quantitatively different from those of MCT. Because of this discrepancy it has been assumed in the past that the two methods are intrinsically different. Instead Szamel [13] has shown that starting from replicated Ornstein-Zernicke equation and closing them with a suitable approximation scheme one can recover exactly the results of MCT. Therefore it can be argued that the two approaches predict the same physics and that discrepancies arise only when inequivalent approximation schemes are used.

Obviously the two methods cannot be completely equivalent. In principle MCT can describe dynamics on both small and large times scale while the replica method is intrinsically a static approach. Therefore it is reasonable that the latter can describe a static (or rather quasi-static) quantity (like the ergodicity-breaking parameter) while it cannot obviously tell anything on short-time dynamics. Previously it was assumed that it could not tell anything on dynamics at all. Instead it has been recently realized [14] that it can be used to characterize the critical behavior on the large time scale of the β regime, *i.e.* the plateau in the time correlators. In order to do so one has to perform an expansion of the equation for the order

parameter q_{ab} around the solution $q_{ab} = q_c$ corresponding to the glassy phase. Independently of the approximate method used in order to obtain the equation it follows that we should get an equation in powers of $q_{ab} = q_c + \delta q_{ab}$ of this form:

$$0 = \tau + m_2 \left(\sum_c \delta q_{bc} + \delta q_{ac} \right) + m_3 \sum_{cd} \delta q_{cd} + w_1 (\delta q^2)_{ab} + w_2 \delta q_{ab}^2 + O(\delta q^3). \quad (1)$$

The various coefficients depend on the given model and on the external parameters, in particular τ vanishes linearly with the external parameters at the critical point and is positive(negative) in the glassy(liquid) phase. In [14] it has been shown that the structure of the above equation determines the behavior of the time correlators near the critical point, in particular the critical behavior of the two-point time correlation $C(t)$ around the plateau value $C_p = q_c$ is given by:

$$\delta C(t) \equiv C(t) - C_p = |\tau|^{1/2} f_{\pm}(t/t^*) \quad t \gg 1, \quad t^* \propto \frac{1}{|\tau|^{1/2a}} \quad (2)$$

where the function f_- has to be chosen in the liquid phase ($\tau < 0$) while the function f_+ has to be chosen in the glassy phase ($\tau > 0$) and t^* is the time scale of the β -regime. The function $f_{\pm}(x)$ obeys the scale-invariant equation:

$$\pm 1 = f_{\pm}^2(x) \left(1 - \frac{w_2}{w_1} \right) + \int_0^x (f_{\pm}(x-y) - f_{\pm}(x)) \dot{f}_{\pm}(y) dy \quad (3)$$

For small values of x both the functions $f_{\pm}(x)$ diverge as $1/x^a$, while for large values of x $f_+(x)$ goes to a constant while $f_-(x)$ diverges as $-x^b$ where the exponents a and b are determined by the so-called parameter exponent λ which turns out to be equal to the ratio w_2/w_1 :

$$\lambda \equiv \frac{w_2}{w_1} = \frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \frac{\Gamma^2(1+b)}{\Gamma(1+2b)} \quad (4)$$

Note the the above results are quite general and the microscopic details of the given model enters only in the actual value of the parameter exponent λ . The above equations are equivalent to those obtained originally in the context of MCT by Götze (see eqs. 22a and 24 [3]) who also recognized their exact nature, *i.e.* the fact that the (model-dependent) value of λ is the only quantity sensitive to the approximation scheme used. Note the this result is another hint at the equivalence between the replica approach and MCT because it shows that their predictions do coincide when they are exact.

The result derived in [14] provides a simple recipe to determine λ within the replica approach: one has to obtain a closed equation for the non-ergodicity parameter using his preferred approximation scheme, expand the equation around the critical value of q_{ab} up to second order as in eq. (1), read the coefficients w_2, w_1 and finally

identify λ with their ratio. This recipe has been applied to a number of mean-field spin-glass models yielding new analytical predictions concerning critical dynamics [15–18]. When there was already an explicit solution of the dynamics the novel computations offered an *a posteriori* validation of the recipe, while in other cases the new predictions were compared with existing numerical simulations. More recently the recipe has been used in the context of the replicated theory of super-cooled liquids based on the HNC approximation [12] and for a wide range of models a (quite reasonable) value of $\lambda \approx .7$ was obtained. In order to show that the replica approach and MCT provide equivalent descriptions of the long-time critical dynamics in the following the recipe will be instead applied to Szamel's approximation scheme.

The issue of the equivalence between the two approaches is not an academic one. On one hand MCT predictions are more accurate at present than those obtained with the HNC approximation, at least in three dimension. On the other hand there are situations in which the replica method may have considerable advantages. Indeed in order to derive many quantities within MCT one takes the large-time limit in the dynamical equations. The replica approach can be viewed as a compact way to describe the algebra of this limit where different replicas correspond to configurations visited dynamically at distant times. Certainly the main open problem concerning MCT is the systematic inclusion of corrections and the role finite-dimensional effects/finite-size effects [19, 20]. The replica approach offers the possibility to study this phenomena in a field-theoretic framework that is definitively simpler than the dynamical one. Surprisingly it has been recently discovered that the loop corrections to this theory can be controlled at all orders and a series of novel unexpected predictions have been obtained [19]. However this study confirmed that the phase transition predicted by MCT is only a dynamical cross-over and therefore the purely static replica approach, as expected, is inconsistent beyond perturbation theory. The theory needs therefore to be supplemented with some form of critical dynamics but unfortunately in spite of important progresses [21] we are still lacking full control of the critical behavior of the dynamical propagators, like the well-known χ_4 susceptibility.

Computation of the parameter exponent Our starting point is the replicated Ornstein-Zernicke equations introduced by Szamel in Ref. [13].

$$\delta n_{ab}(\mathbf{k}) = n^2 c_{ab}(\mathbf{k}) + n \sum_c c_{ac}(\mathbf{k}) \delta n_{cb}(\mathbf{k}) \quad (5)$$

where $\delta n_{ab}(\mathbf{k})$ is the Fourier transform of $\delta n_{ab}(\mathbf{r}_1, \mathbf{r}_2) = n_{ab}(\mathbf{r}_1, \mathbf{r}_2) - n^2$, *i.e.* the non trivial part of the two-particle density, n being the total particle density. The above equations can be closed provided an expression for the direct correlations $c_{ab}(\mathbf{k})$ in terms of the $\delta n_{ab}(\mathbf{k})$ itself is given. The single replica correlation functions are associated by the definition to the static structure factor

according to:

$$\delta n_{aa}(\mathbf{k}) = n(S(k) - 1) \quad (6)$$

Under the assumption of Replica-Symmetry (RS) the equation for the diagonal components factorizes from that of the off-diagonal components in the limit $m \rightarrow 1$ and equation (5) reduces to

$$\delta n_{aa}(\mathbf{k}) = n^2 c_{aa}(\mathbf{k}) S(k) \quad (7)$$

the above equation combined with the definition (6) can be seen as a definition of the direct correlation function:

$$c_{aa}(\mathbf{k}) = c(k) \equiv \frac{S(k) - 1}{n S(k)} \quad (8)$$

Indeed the static structure factor is usually considered an input in MCT computations. Szamel has shown that that making an appropriate set of approximations in the context of the Yvon-Born-Green hierarchy one obtains an expression for the off-diagonal direct correlation identical to Sjogren's vertex [7]. More precisely if we define the replicated non-ergodicity parameter $f_{ab}(\mathbf{k})$ according to:

$$\delta n_{ab}(\mathbf{k}) = n f_{ab}(k) S(k) \quad (9)$$

we obtain the expression:

$$c_{ab}(\mathbf{k}) = \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) f_{ab}(q) f_{ab}(p) \quad (10)$$

where the function $G(\mathbf{k}, \mathbf{q}, \mathbf{p})$ has precisely Sjogren's form:

$$G(\mathbf{k}, \mathbf{q}, \mathbf{p}) = \frac{1}{2k^2 (2\pi)^3} \delta[\mathbf{k} - \mathbf{p} - \mathbf{q}] S(q) S(p) (\hat{\mathbf{k}} \cdot [\mathbf{q} c(q) + \mathbf{p} c(p)])^{20} = -\frac{f(k)}{(1-f(k))} - \delta q_{ab}(\mathbf{k}) + (1-f(k))(\delta q^2)_{ab}(\mathbf{k}) + \quad (11)$$

Now we multiply eq. (5) by the inverse of the matrix $nI + \delta n_{ac}(\mathbf{k})$, (I is the identity in replica and momentum space), and we use the above equation to obtain:

$$\left(\frac{\delta n}{nI + \delta n} \right)_{ab}(\mathbf{k}) = n \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) f_{ab}(q) f_{ab}(p). \quad (12)$$

In order to perform an expansion around the RS solution we rewrite:

$$\delta n_{ab}(\mathbf{k}) = \delta n_{ab}^{RS}(\mathbf{k}) + nS(k)(1-f(k))^2 \delta q_{ab}(\mathbf{k}) \quad (13)$$

where $\delta n_{ab}^{RS}(\mathbf{k})$ has a RS structure

$$\delta n_{aa}^{RS}(\mathbf{k}) = n(S(k) - 1) \quad (14)$$

$$\delta n_{ab}^{RS}(\mathbf{k}) = n f(q) S(q) \quad a \neq b \quad (15)$$

The factor $nS(k)(1-f(k))^2$ has been used in the definition in order to make contact with ref. [3]. According to [14] we have to expand equation (12) in powers of $\delta q_{ab}(\mathbf{k})$ assuming $\delta q_{aa}(\mathbf{k}) = 0$.

In order to perform the expansion we first note that the r.h.s. (where the crucial approximations are made)

has a simple diagonal structure in replica space and only the l.h.s. has a non-trivial structure. The zero-th order term can be computed using standard properties of replica symmetric matrices and leads after some algebra to:

$$\left(\frac{\delta n}{nI + \delta n} \right)_{ab}(\mathbf{k}) = \frac{f(k)}{S(k)(1-f(k))} + O(\delta q), \quad a \neq b \quad (16)$$

In order to compute the next terms we note that in a power series expansion they reduce to the computation of objects of three possible forms: 1) $(\delta n_{RS})_{ab}^r$, 2) $(\delta n_{RS}^r \delta q \delta n_{RS}^s)_{ab}$ and 3) $(\delta n_{RS}^r \delta q \delta n_{RS}^s \delta q \delta n_{RS}^t)_{ab}$ for general integer r, s, t (all products and powers are intended in matrix sense and we have not written explicitly the replica and Fourier indexes). At linear order the expansion generates terms of three types: $m_1 \delta q_{ab}$, $m_2 \sum_c (\delta q_{ac} + \delta q_{bc})$ and $m_3 \sum_{cd} \delta q_{cd}$. However, as explained in Ref. [14], only the first one, that depends simultaneously on both indexes a and b , is relevant for the dynamics [23]. The computation of this term is simpler than the others because since both indexes must be present every time we have a multiplication by $\delta n_{RS}(k) = n(S(k)(1-f(k)) - 1)\delta_{ab} + f(k)S(k)$ we are forced to take the term with the Kronecker delta. The same observation is correct also at quadratic order in $\delta q(\mathbf{k})$, i.e. for the term $\sum_c \delta q_{ac}(\mathbf{k}) \delta q_{cb}(\mathbf{k})$. Therefore in order to compute the terms relevant for the dynamics we can replace the much simpler form $\delta n_{ab}(\mathbf{k}) = n(S(k)(1-f(k)) - 1)\delta_{ab} + nS(q)(1-f(q))^2 \delta q_{ab}(\mathbf{k})$ into the l.h.s. of eq. (12). Considering only the terms relevant for the dynamics, the final expression is:

$$\begin{aligned} & -\frac{f(k)}{(1-f(k))} - \delta q_{ab}(\mathbf{k}) + (1-f(k))(\delta q^2)_{ab}(\mathbf{k}) + \\ & + nS(k) \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) f(q) f(p) + \\ & + 2nS(k) \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) (1-f(q))^2 \delta q_{ab}(\mathbf{q}) f(p) + \\ & + nS(k) \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) \times \\ & \times (1-f(q))^2 (1-f(p))^2 \delta q_{ab}(\mathbf{q}) \delta q_{ab}(\mathbf{p}) \end{aligned} \quad (17)$$

The equation at zero-th order determines the non-ergodicity parameter while the next orders determine the critical temperature and the parameter exponent. The critical temperature is fixed by the condition that there is a direction for $\delta q_{ab}(k)$ such that the coefficient of the linear term vanishes. This corresponds to the condition that the following linear operator has a zero eigenvalue $\mu = 0$:

$$\begin{aligned} \mu \xi_\mu^R(k) &= -\xi_\mu^R(k) + 2nS(k) \int d\mathbf{q} d\mathbf{p} G(\mathbf{k}, \mathbf{q}, \mathbf{p}) \times \\ &\times (1-f(q))^2 \xi_\mu^R(q) f(p) \end{aligned} \quad (18)$$

where $\xi_\mu^R(k)$ is the right eigenvector corresponding to eigenvalue μ . Note that this condition is equivalent to

the condition that the derivative with respect to $f(q)$ of the zero-th order term vanishes and therefore marks the point in temperature where a non vanishing $f(q)$ appears. This equivalence between two different conditions is a consequence of the $m \rightarrow 1$ limit and of the fact that in this limit the so-called replicon and longitudinal eigenvalue are degenerate leading to many non trivial features concerning fluctuations [19]. The behavior at the critical point is controlled by the critical eigenvector $\xi_\mu^R(k)$ and we may write at leading order:

$$\delta q_{ab}(k) = \delta q_{ab} \xi_0^R(k) . \quad (19)$$

Note that the above definition must be supplemented with a normalization condition on the eigenvector, in order to make contact with ref. [3] we adopt the following normalization for the right and left eigenvectors:

$$\int d\mathbf{k} \xi_0^L(k) \xi_0^R(k) = 1 , \int d\mathbf{k} \xi_0^L(k) (1-f(k)) \xi_0^R(k) \xi_0^R(k) = 1 .$$

Multiplying the quadratic part of equation (17) by the left eigenvector $\xi_0^L(k)$ and integrating we obtain the following expression:

$$\left[\int d\mathbf{k} \xi_0^L(k) (1-f(k)) \xi_0^R(k) \xi_0^R(k) \right] (\delta q_{ab})^2 + n \left[\int d\mathbf{k} d\mathbf{q} d\mathbf{p} \xi_0^L(k) S(k) G(\mathbf{k}, \mathbf{q}, \mathbf{p}) (1-f(q))^2 (1-f(p))^2 \xi_0^R(q) \xi_0^R(p) \right] \delta q_{ab}^2 , \quad (20)$$

Note that the first coefficient is fixed to one by the normalization. According to [14] the parameter exponent is given by the ratio between the two quadratic coefficients leading to:

$$\lambda = n \int d\mathbf{k} d\mathbf{q} d\mathbf{p} \xi_0^L(k) S(k) G(\mathbf{k}, \mathbf{q}, \mathbf{p}) \times (1-f(q))^2 (1-f(p))^2 \xi_0^R(q) \xi_0^R(p) \quad (21)$$

Identifying the two-point vertex in [3] as $V^{(2)}(\mathbf{k}; \mathbf{q}, \mathbf{p}) = 2nS(k) G(\mathbf{k}, \mathbf{q}, \mathbf{p})$ we see that that eqs. (18) and (21) are respectively equivalent to eq. (13a) and (15a) in [3].

Conclusions The recipe of [14] has been applied to the approximation scheme proposed by Szamel in order to show that the replica approach gives a characterization of the β -regime of super-cooled liquids that is equivalent to the one of MCT both qualitatively and quanti-

tatively, provided the same approximations are used. A novel result obtained within the replica approach is that an equation like (1) follows from a replicated free energy [14]. This allows to connect the coefficients w_1 and w_2 with two six-point susceptibilities ω_1 and ω_2 which are in principle measurable in a quasi-static framework. According to [14] their ratio is equal to λ and therefore eq. (21) fixes their value up to an unknown constant. The computation of this constant is left for future work. In the present framework it could be determined considering the response of the non-ergodicity parameter to a random pinning field along the lines of [22].

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